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Patent- og Varemærkestyrelsen
Økonomi- og Erhvervsministeriet

24 November 2003

A handwritten signature in black ink, appearing to read "Pia Høybye-Olsen".

Pia Høybye-Olsen

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**Process for Removal of SO₂ from
Off-Gases by Reaction with H₂O₂**

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The present invention relates to a process for removal of SO₂ from off-gases by reaction with H₂O₂.

It has been known for more than 30 years that SO₂, as described in the publication 2164e from Lurgi/Südchemie AG, August 1989, can be removed from off-gases by contacting the off-gas in an absorption tower with circulating solution of dilute sulphuric acid containing H₂O₂, whereby SO₂ is dissolved and oxidised to H₂SO₄ in the solution. The 5 circulating solution typically contains 30-60% H₂SO₄ and 0.1-0.5% H₂O₂. The absorption is typically carried out at a temperature of 50-80°C of the circulating solution. H₂O₂ is added either as a concentrated aqueous solution of H₂O₂ to the circulating acid, or it is produced by electrolysis of 10 a side stream of the circulating acid. The produced acid is added to the circulating acid. The produced acid is drawn off from the circulating acid.

The known process usually requires installation of a low 20 velocity aerosol filter downstream of the absorption tower to remove sulphuric acid aerosol (acid mist) in order to meet acid mist emission regulations requiring less than about 5 mol ppm H₂SO₄ in the stack gas. Fine acid mist (aerosol) that may be present in the off-gas is not removed 25 efficiently in the absorption tower. Fine acid mist is also formed in the absorption tower itself by reaction between SO₂ and H₂O₂ vaporised from the absorbing liquid.

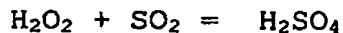
It is a disadvantage of the known process that it requires 30 installation of both an absorption tower and a low velocity mist filter.

In the process according to the present invention, SO₂ in off-gases is removed by reaction with H₂O₂ without the use of an absorption tower by spraying a solution of H₂O₂ in water or dilute sulphuric acid into the off-gas upstream of a low velocity aerosol filter or wet electrostatic precipitator (WESP).

A preferred embodiment of the invention is shown in Figure 1. A solution of 0.1-30% H₂O₂ in line 1 is sprayed by the spray nozzles 3 into a stream of off-gas in line 2 containing typically between 100-1000 ppm SO₂ and having a temperature typically in the range of 50-120°C. The nozzles are placed in duct 4, so that the spray is evenly distributed in the gas stream upstream of the mist filter 5 in which the gas is passed in parallel through a number of low velocity filter candles 6. Even distribution of the droplets in the gas is desirable for the process, and the most even distribution of the droplets is achieved by using air-atomising nozzles producing very small droplets. The H₂SO₄ formed in the process accumulates in the filter elements or candles from which it is drained off through line 7. Most or all of the mass of the droplets evaporate before the gas enters the filter candles, whereby most of the H₂O₂ evaporates and reacts in the gas phase under formation of sulphuric acid aerosol. However, it is not necessary that the droplets are completely evaporated before the gas enters the filter elements. The reaction between SO₂ and H₂O₂ will be completed and the thermal equilibrium will be established in the mist filter elements without decreasing the efficiency of the SO₂-removal.

Thus the injection of the aqueous solution H_2O_2 serves two purposes:

5 Firstly, it adds to the off-gas the amount of H_2O_2 , which is required for achieving the desired conversion of SO_2 into H_2SO_4 by the reaction



10 Most of the conversion takes place by reaction in the gas phase between SO_2 and vaporised H_2O_2 under formation of acid mist, or between SO_2 and H_2O_2 dissolved in the droplets. The reaction is completed in the aerosol filter in which remaining SO_2 is absorbed and reacts with remaining H_2O_2 contained in the dilute sulphuric acid wetting the fibre material.

20 Secondly, the water comprised in the solution cools off the off-gas in line 2 by evaporation of the droplets, whereby the off-gas is cooled off to a desired temperature of the filter elements or candles typically to a temperature between 50°C and 70°C. The concentration of H_2SO_4 in the produced acid will be the equilibrium concentration of H_2SO_4 at the actual temperature and H_2O partial pressure in the gas phase.

25 Up to 98% SO_2 -removal can be achieved at, typically, about 95% utilisation of the H_2O_2 .

Example

An off-gas stream of 1000 Nm³/h at 100°C contains 200 ppm SO₂ + 10% H₂O and has a temperature of 100°C. 96% SO₂ removal is desired. The aerosol filter is designed for operation at maximum 70°C. Operation at 67-70°C is chosen in order to achieve the highest possible acid strength and low content of remaining H₂O₂ in the produced acid.

The process is conducted as follows: 15.7 kg/h water containing 2.0 wt% H₂O₂ is injected into the off-gas, whereby the off-gas is cooled to 65-70°C in thermal equilibrium. The mist filter is 75 mm thick and has a flow area of 2.5 m². The diameter of the fibres is about 8 µm. Experiments carried out under these conditions show that about 96% of the SO₂ is removed under production of 1.7 kg/h 50% H₂SO₄ with about 500 ppm H₂O₂. The treated gas contains less than 2 ppm H₂SO₄, and the content of H₂O₂ is below the detection limit.

CLAIMS

1. A process for removal of SO₂ in off-gases having a temperature of 30-150°C and containing 0.001-1 vol% SO₂ in which the SO² is oxidised to H₂SO₄ by spraying an aqueous solution of H₂O₂ into the off-gas upstream of an aerosol filter removing the produced sulphuric acid from the off-gas.
- 10 2. A process as in claim 1 in which the off-gas is cooled by evaporation of the water comprised in the solution being sprayed into the off-gas upstream of the filter.
- 15 3. A process as in claim 1 in which a wet electrostatic separator is used in place of an aerosol filter.

ABSTRACT

A process for removal of SO₂ in off-gases having a temperature of 30-150°C and containing 0.001-1 vol% SO₂ in which 5 the SO² is oxidised to H₂SO₄ by spraying an aqueous solution of H₂O₂ into the off-gas upstream of an aerosol filter removing the produced sulphuric acid from the off-gas.

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21 DEC. 2002

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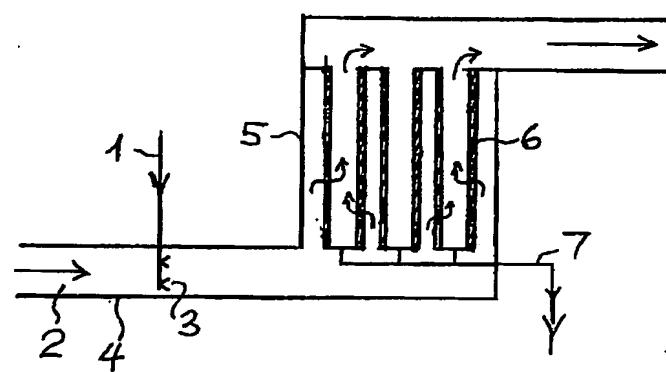


Fig 1